

The theory of surface spin combustion

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Abstract - Experimental investigation of condensed system combustion has resulted in discovery of a new spin regime of reaction zone propagation. In the spin burning the small reaction spot moving along the surface of the cylindrical sample is observed.

A brief review of experimental, numerical, and analytical studies is presented. It leads to a new concept of two types of spin referred to strong and weak spins. The former is a very pronounced nonlinear phenomenon obtained experimentally. The latter represents only a result of theoretical studies performed mainly in the framework of the bifurcation theory and never observed in experiment. There is a significant discrepancy between quantitative characteristics of the two types of spin.

An approximate analytical method for describing strong surface spin combustion is formulated in the paper. It is assumed in a two-dimensional model that both unburned fuel and combustion products are solid and the condensed phase exothermic reaction occurs at the cylindrical surface, with the combustion centre penetrating into the specimen to a small depth. So as the spin spot motion is almost perpendicular to the vector of the combustion wave mean velocity the two-dimensional problem can be reduced to a simple one-dimensional one which is similar to the problem of combustion with heat losses.

The conditions of onset of the spin combustion regime are found out. Explicit analytical expressions for various quantities of interest, including the propagation spin velocity, the wave mean velocity, and the temperature profile are given. They depend on cylinder diameter and heat losses. As the diameter grows, two or more centres of chemical reaction appear (multi-centre spin) at the cylinder surface. Wherever possible, theoretical results are compared with experimental data.

INTRODUCTION

For the first time spin burning has been observed by Merzhanov and co-workers (refs.1,2) in studying the burning of porous metallic specimens in gaseous oxidizer. On igniting the cylindrical sample glowing spot of small diameter appears on its lateral surface and spirals along the surface. The spot leaves a bright trace which gradually merges with a similar trace of the previous turn. Thus, only thin lateral surface layer is consumed at the first stage of combustion; the inner part of the sample either does not burn out or reacts partly or completely after passing the surface combustion front.

Similar regimes of reaction zone propagation have been obtained in studying gasless combustion (refs.3,4). In this case a cylindrical specimen burns over its volume. The combustion front propagates as a plane moving around the cylinder axis. The experimental evidence for multi-spin regimes has also been found (refs.4-6). In this case, under certain conditions, several luminous spots spiral down the lateral surface of the sample.

The spin combustion as well as other nonlinear nonsteady phenomena observed in combustion of systems yielding solid-phase products are related to the existence of stability boundary of a steady-state flat reaction wave. To investigate these combustion modes one should study propagation regimes in the instability domain. Recently, the author has published a review (ref.7) to provide readers with numerous experimental, analytical, and numerical results obtained in this field of combustion science.

We present first the most important results of a linear stability analysis of stationary solids combustion. It has been shown by Makhviladze and Novozhilov (ref.8), that when a parameter related to the activation energy exceeds a critical value, instability of stationary regime with multidimensional perturbations arises more readily than it does in the one-dimensional case and the stability failure is of oscillatory nature.

On describing the nonsteady modes of combustion on the unstable side of the neutral stability boundary a nonlinear analysis is needed to calculate finite amplitudes perturbations. Rather a great number of papers is devoted to studying this problem with the help of bifurcation theory. This theory is concerned with the determination of the amplitudes of perturbations only in the vicinity of the neutral stability boundary where the deviation of the burning regime from the steady-state can be treated as a small correction. We call this type of spin the weak one. There is no experimental evidence for the existence of the weak spin.

The real spin, observed experimentally, is a very pronounced non-linear combustion regime and its characteristics differ drastically from those of the weak spin. This type of spin is referred to as a strong one.

An approximate theory of strong surface spin combustion regimes is presented in the paper.

STABILITY BOUNDARY OF STEADY-STATE PLANAR FRONT

Several analytical and numerical studies originated from Zeldovich's work (ref.9) were undertaken in order to shed some light on the facts of appearance of one- and two-dimensional instabilities in solid propellant and gasless combustion (refs.10-13). The problem of the stability of stationary combustion regime of condensed systems with respect to flame front deformations has been formulated by Makhviladze and the author (ref.8). Since the full analytical solution of this problem is not possible the approximation of infinitely thin chemical conversion zone was adopted for the first time. The analysis is connected with investigation of the thermal diffusion stability of a laminar gas flame (ref.14).

In terms of this approach one considers the zone of chemical conversion in original substance very thin in comparison with the width of the preheating zone, and regards it as a surface separating the initial substance from the reaction products. As a result, the problem is incomplete and, therefore, the approximation of infinitely narrow reaction zone necessitates a "closure" hypothesis for the rate of unsteady-state burning.

Further on we will assume that the linear burning rate u depends exclusively on temperature in the reaction zone, T_b . Then the variation in burning rate with temperature can be described in a linear approximation by the coefficient

$$k = (T_b^0 - T_a) d \ln u^0 / d T_b^0 \quad (1)$$

whose explicit expression depends on the type of steady-state burning law $u^0(T_b^0)$. Here T_a is the initial temperature, and the quantities pertaining to the stationary combustion regime are labelled with zero superscript. In this description the results of the stability analysis should have more wide applicability range than they do in the case of the Arrhenius reaction rate temperature dependence.

Assuming that x is the coordinate normal to the unperturbed flame front we can take a small perturbation in the form

$$\delta T \sim A(x) \exp(\Omega t + iKy) \quad (2)$$

where y is the coordinate along the front and t is the time. The values K and Ω are the wave number and complex frequency, respectively.

In the following we will use nondimensional variables and parameters. Let κ denote thermal diffusivity then $s = 2K\kappa/u^0$ and $\omega = \Omega\kappa/(u^0)^2$ are the nondimensional wave number and frequency respectively.

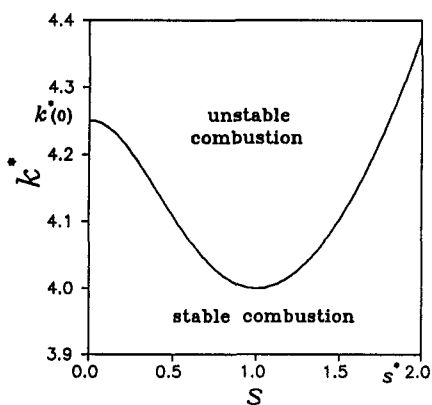


Fig. 1. Stability boundary of steady - state combustion regimes. The meaningful points: $s^* = (1 + 5^{1/2})^{1/2}$, $k^*(0) = k^*(s^*) = 2 + 5^{1/2}$, $k^*(1) = 4$

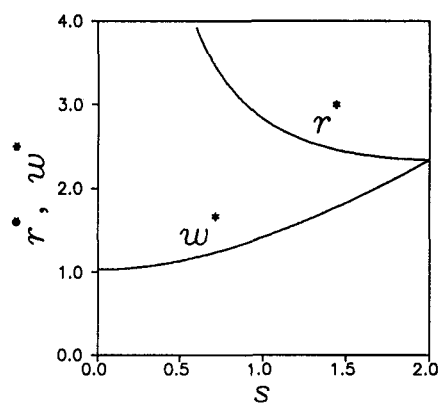


Fig. 2. Frequency and the ratio of velocity components at the stability boundary versus wave number.

A perturbation theory analysis applied to the combustion stability problem gives the following result concerning the stability boundary (ref.8). The steady-state regime is stable at $k < k^*(s)$ where

$$k^*(s) = \left(4 + 3s^2 + \sqrt{(4 + 3s^2)^2 + 4(1 + s^2)^3} \right) / 2(1 + s^2) \tag{3}$$

Figure 1 shows the domains of stability and instability separated by the curve $k^*(s)$. The frequency at the stability boundary is purely imaginary, i.e. the stability loss is of oscillatory nature. It can be easily calculated using the expression:

$$(\omega^*)^2 = (1 + s^2)k^*(s)/4 \tag{4}$$

Equations 2 - 4 allow one to find the velocity of small perturbation along the flame front $v = |\Omega|/K$. It is useful to introduce the ratio of two velocity components at the stability boundary $r^* = v/u^0$. Then we have

$$r^* = [(1 + s^2)k^*(s)]^{1/2}/s \tag{5}$$

Graphs of the functions ω^* and r^* are given in Fig.2.

Assuming $s = 0$ in Eq.3, we find that the region of instability for one-dimensional perturbations is located at $k > k^*(0)$, where $k^*(0) = 2 + 5^{1/2}$. The minimum value of the parameter characterizing the stability, $k_{min}^* = 4$, is attained for $s = 1$. This indicates that two-dimensional perturbations could be more dangerous than one-dimensional ones.

However within the framework of the adopted approximation this conclusion is not rigorous enough. Really the analysis neglects the temperature variation across the reaction zone ΔT (which is of the order of magnitude RT_b^2/E) in comparison with the characteristic temperature range $T_b^0 - T_a$. Thus, the value $\Delta T/(T_b^0 - T_a) \sim 1/k$ is considered very small. It is known that near the stability boundary $k \approx 4$, therefore, the error introduced into the calculation is of the order of ten per cent. At the same time, the decrease in parameter k^* with the variation of s from zero to unity is only six per cent. Consequently the inference about the presence of a minimum on the curve $k^*(s)$ is not rigorous because the effect is of the order of the error permissible in calculation (refs.11,15).

The author and co-workers (refs.16,17) examined numerically the problem of the stability for gasless system stationary combustion taking into account the finite thickness of the reaction zone. The cases of various order chemical reactions and a broad zone were studied, and the stability limit was found in the linear approximation. It was really shown that two-dimensional perturbations more dangerous than one-dimensional ones. Interpolation formulae have been given for the parameters of the system and for the frequency of the perturbations versus the wavelength of the perturbation at the stability limit.

WEAK AND STRONG SPINS

In previous section we considered the combustion wave propagation through infinite two-dimensional flat layer. Thus a continuous spectrum of transverse wave numbers is possible in the problem. However in a typical experiment a combustion wave propagates through a long enough sample of finite transverse dimensions and the boundary conditions discretize the admissible transverse wave numbers. Perturbation modes in such a case are eigenfunctions of the Laplace operator related to the cross-section of the sample.

Several theoretical works (refs.18-25) have discussed the stability problem using two - and three - dimensional approaches. It has been shown that in cylindrical geometry transition to spin combustion can occur as a result of the loss of the stability of a stationary planar wave. To demonstrate the existence of these solutions a nonlinear analysis is necessary. However, the main characteristics of the phenomena may be obtained by a linear analysis presented above. The aim of this section is to show that spin combustion waves given by the bifurcation theory differ drastically from those observed experimentally.

First of all, it should be emphasized that the flame front in the bifurcation theory is considered to be a stationary plane front with a small time-dependent correction. Thus, in such a case the front is continuous and only slightly differs from the steady-state one. To the contrary, a strong spin both in surface combustion and in volume combustion has a disrupted front (reaction spot).

To compare quantitative features of the two types of spin combustion we shall discuss a simple example of a cylindrical specimen with surface combustion. This problem arises in the case of surface filtration combustion. Let a cylinder surface be cut along the longitudinal axis and unrolled. As a result, we receive an infinitely long strip with periodical condition on its boundaries.

The values of the admissible wave numbers can be obtained from the condition $\pi d = m\lambda_m$ where d is the cylinder diameter, $\lambda_m = 2\pi/K_m$ is the wave length and m is any positive integer. Thus, we have $s_m = 4m\kappa/du^0$ as possible dimensionless wave numbers.

The set of different combustion modes excited is defined by the ratio κ/du^0 . If $d \ll \kappa/u^0$ one has $s_m \gg 1$ and the flat pulsation regime is realised. If the ratio κ/du is small the modes with large wave numbers are excited first. Two examples of possible sets of discrete mode eigenvalues are shown in Figs. 3 and 4.

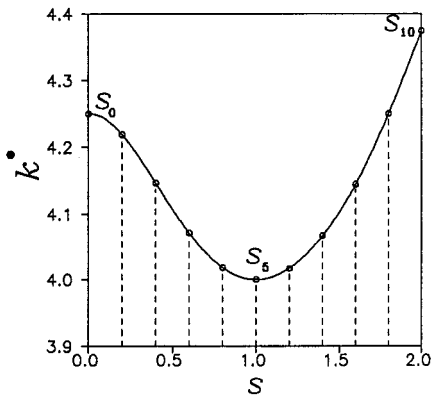


Fig. 3. The location of discrete modes for $d = 20 \kappa/u^0$. The fifth mode first loses stability with increasing k .

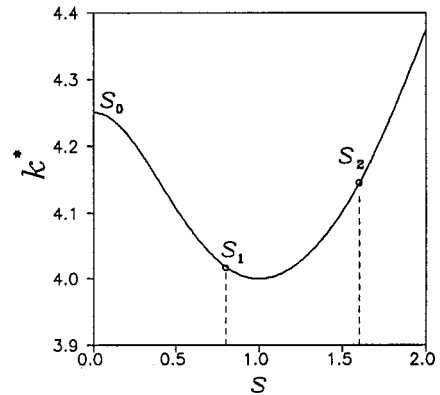


Fig. 4. The location of discrete modes for $d = 5 \kappa/u^0$. The first mode is the most dangerous one.

The one - head spin is the first one entering the instability domain (by increasing the instability parameter k) if

$$k_1^*(s) < k^*(0) \text{ and } k_1^*(s) < k_m^*(s) \text{ at any } m - \text{value.}$$

To the contrary, if

$$k^*(s_1) > k^*(0) \quad \text{or} \quad k^*(s_1) > k^*(s_m)$$

the first excited mode is the plane one or the multi-head spin. Since $s_1 = 2s_2$ we can easily obtain from Eq.3 that the interval in which the one-head spin is excited first is $0.67 < s_1 < 1.8$. It follows from Eq.5 and Fig.2 that the ratio of the velocity components for the one-head spin in the bifurcation

theory is $2.4 < r^* < 3.6$. These values are not large enough to explain the experimental data. In fact, spin combustion experiments (refs.1-4,26-29) show that the ratio of velocity components v/u is in the range from 10 to 30.

Lastly, the range of s values at which the one-head spin arises corresponds to very small cylinder diameters and one can derive that the weak spin may be realized only at $2.2\kappa/u^0 < d < 6.0\kappa/u^0$. Real strong spin experiments deal with specimens of much larger diameter.

A mathematical model of spin burning has been suggested and numerically realized by Shkadinsky and co-workers (refs.30,31). It was assumed that a condensed phase first-order exothermic reaction occurs at a cylindrical surface. Spin burning has been numerically obtained in the instability domain of steady-state combustion only at very small specimen diameters ($du^0/\kappa < 5, v/u^0 < 3.2$). However real experiments are run with specimens of much larger diameters. Hence, the problem of spin combustion at real diameter values cannot be regarded as resolved.

APPROXIMATE THEORY OF STRONG SPIN

An approximate analytical approach to the investigation of surface spin combustion (ref.32) is presented below.

The scheme of the process is presented by Fig.5. The process is considered to occur in the infinite x - direction strip of width $l = \pi d$ which is a surface evolvent of a cylinder of diameter d . The coordinate system is chosen so that the reaction zone is fixed while substance moves in x - and y - directions with velocities u and v , respectively. It is assumed that the reaction occurs in an infinitely thin zone, the burning rate being known as a function of temperature of this zone. Dash line in Fig. 5 indicates the boundary between burned and unburned parts of the sample.

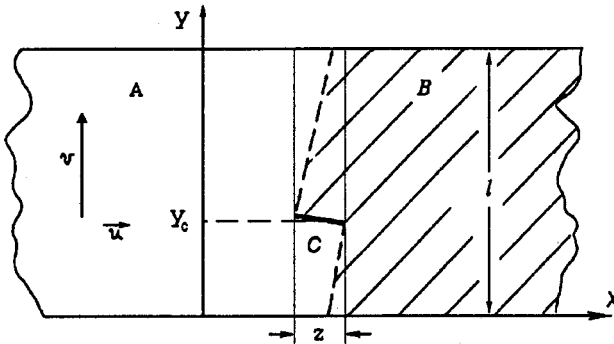


Fig.5. Sketch of a burning strip:
 (A) virgin substance,
 (B) combustion products,
 (C) flame front.

The width of combustion zone is equal to the spin step z . The law of mass conservation gives $lu = zv$. At $l \gg z$, the temperature is assumed to be constant over the thin substance layer of width z . This layer may be considered to be a reactor receiving x - directed flux from unburned sample.

The processes within this layer can be described by the thermal conductivity equation

$$\kappa \frac{d^2 T}{dy^2} - v \frac{dT}{dy} + q_1 - q_2 - q_3 - q_4 = 0, \tag{6}$$

where

- $q_1 = \frac{u}{z} (T_a - T)$ - convective energy flux,
- $q_2 = a \frac{\kappa}{z^2} (T - \frac{T}{a})$ - heat losses into the green mixture,
- $q_3 = a_1 \frac{\kappa}{z^2} (T - T_b)$ - heat losses into the combustion products,
- $q_4 = \frac{\kappa}{h^2} (T - T_a)$ - heat losses into the surroundings.

The values a and a_1 are the corresponding coefficients of heat losses. The effective thickness of the layer in radial direction h is introduced to characterize the heat losses in that direction.

The combustion zone can be conveniently placed at $y = 0$, the corresponding boundary conditions being the following

$$T|_{y=l} = T|_y, \quad \kappa \frac{dT}{dy} \Big|_{y=l} - \kappa \frac{dT}{dy} \Big|_{y=0} = v(T_b - T_a) \tag{7}$$

The combustion zone thickness should be taken to be of the same order of magnitude as the thickness of heated layer along x - axis, $z = b\kappa/u$, where b is a coefficient of the order of unity. From the last expression and the relationship $z = ul/v$ one obtain $z = (b\kappa l/v)^{1/2}$.

Assuming the Arrhenius dependence of burning rate on temperature, the burning rate relates to the combustion zone temperature as follows: $v = A \exp(-E/RT)$. Then, let's introduce the dimensionless variables and parameters:

$$\begin{aligned} V &= v/u^0, & U &= u/u^0, & L &= lu^0/\kappa, & H &= hu^0/\kappa \\ \Theta_c &= T_c/T_b, & \Theta_a &= T_a/T_b \end{aligned} \quad (8)$$

where $u^0 = A \exp(-E/RT_b)$ is the burning rate corresponding to the temperature in the reaction zone T_b . The set of equations (6,7) can be easily solved to give the expression for temperature in the combustion zone

$$\Theta_c = \frac{(1 + a/b + L/VH^2)\Theta_a + a_1/b}{1 + (a + a_1)/b + L/VH^2} + \frac{(1 - \Theta_a)(e_2 - e_1)}{R(1 - e_1)(e_2 - 1)} \quad (9)$$

where

$$\begin{aligned} e_1 &= \exp[-(R + 1)LV/2], & e_2 &= \exp[(R - 1)LV/2], \\ R &= [1 + 4(b_1 + a + a)/bLV + 4/H^2V^2]^{1/2} \end{aligned}$$

The expression (9) and the relationship between reaction rate and temperature in the reaction zone

$$V = \exp[k(1 - 1/\Theta_c)/(1 - \Theta_a)], \quad k = (1 - \Theta_a)E/2RT_b \quad (10)$$

allow one to obtain all the characteristics of spin combustion: $V(L)$, $U = (b/VL)^{1/2}$, and $V/U = (LV/b)^{1/2}$.

It is worth to point out that the model presented above is very similar to the simplest theory (ref.33) of critical diameter of combustion. However, several distinctive features should be mentioned. They are, firstly, the condition of cyclic recurrence (7) which determines the temperature in combustion zone and, secondly, the fact that heat losses are connected not only with external conditions (through parameter h) but also with internal characteristics of the process, namely, with the burning rate along x - axis conforming to the burning rate in perpendicular direction.

The relative width of spin step b as well as the heat loss coefficients a and a_1 depend on real temperature profile in x - direction. If it is treated as the Michelson profile corresponding to the burning rate u one can obtain for the forenamed quantities

$$a = (e - 1)^{-1}, \quad a_1 = e^{-1}, \quad b = 1.$$

These values have been used in calculations and the results are given below. Figures 6 and 7 show the dependence of the transverse burning rate and the ratio of transverse and longitudinal rates on sample diameter at different values of stability parameter and at different intensities of heat losses to environment. The existence of spin limits on both diameter and heat losses and the values of V/U are in agreement with experiment.

The presented model of spin combustion results in theoretical corollary that the ratio of squared velocity along the axis of cylinder and the frequency of spin must be constant and of the same order of magnitude as the thermal diffusivity value. The dependences of burning rate u and frequency ν on sample diameter were reported by Filonenko (ref.26). He found that the frequency decreased by a factor of 3.5 (from 0.7 to 0.2 s^{-1}) and velocity decreased by a factor of 2 (from 0.14 to 0.07 cm/s) when specimen diameter increased from 0.8 cm to 1.8 cm . Meanwhile, the ratio u^2/ν , calculated from experimental data remained fairly constant and was equal to $2.4 \cdot 10^{-2} cm^2/s$. It is easy to recognize that the value of thermal diffusivity is of the same magnitude for typical combustion systems (refs.34-36).

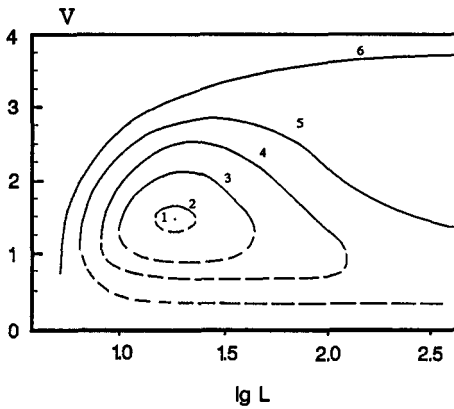


Fig. 6. The dependencies of transverse spin velocity on cylinder perimeter for $k = 5$, $\Theta_a = 0$, and various heat losses.

1. $H = 4.50$; 2. $H = 4.55$; 3. $H = 4.80$;
4. $H = 6.00$; 5. $H = 8.00$; 6. $H = \infty$.

Dashed curves show unstable solutions.

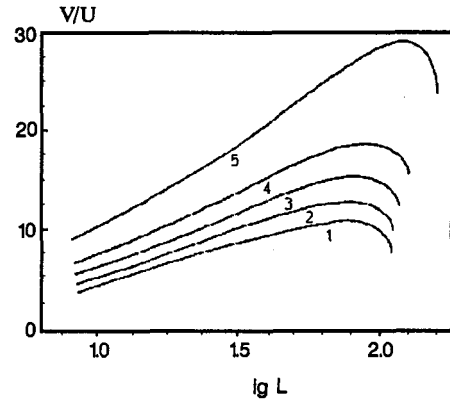


Fig. 7. The ratio of two spin velocity components versus cylinder perimeter for $H = 6$, $\Theta_a = 0$, and various stability parameters.

1. $k = 5$; 2. $k = 6$; 3. $k = 7$;
4. $k = 8$; 5. $k = 10$.

It should be mentioned in conclusion that conditions of existence and characteristics of multi-head combustion spins can be revealed quite easily within the framework of the presented model. It can be shown that

$$V_n(L) = V_1(L/n)$$

where V is the dimensionless velocity of n - head spin.

REFERENCES

1. A.G.Merzhanov, A.K.Filonenko, and I.P.Borovinskaya, *Dokl. Akad. Nauk SSSR*, **208**, 892 - 894 (1973).
2. A.G.Merzhanov, and I.P.Borovinskaya, *Combust. Sci. Technol.*, **10**, 195 - 201 (1975).
3. Yu.M.Maksimov, A.T.Pak, G.V.Lavrenchok, Y.S.Nayborodenko, and A.G.Merzhanov, *Fiz. Goren. Vzryva.*, N° 3, 156 - 159 (1979).
4. Yu.M.Maksimov, A.G.Merzhanov, A.T.Pak and M.N.Kuchkin, *Fiz. Goren. Vzryva.*, N° 4, 51 - 58 (1981).
5. A.G.Strunina and A.V.Dvoryankin, *Dokl. Akad. Nauk SSSR*, **260**, 1185 - 1188 (1981).
6. A.V.Dvoryankin, A.G.Strunina and A.G.Merzhanov, *Fiz. Goren. Vzryva.*, N° 2, 10 - 16 (1982).
7. B.V.Novozhilov, *Pure and Appl. Chem.*, **64**, 955 - 964 (1992).
8. G.M.Mackviladze and B.V.Novozhilov, *Zhurn. Prikl. Mekhaniki i Tekhn. Fiziki*, N° 5, 51 - 59 (1971).
9. Ya.B.Zeldovich, *Zhurn. Eksp. i Teoret. Fiziki*, N° 11-12, 498 - 524 (1942).
10. B.V.Novozhilov, *Zhurn. Prikl. Mekhaniki i Tekhn. Fiziki*, N° 4, 157 - 160 (1965).
11. B.V.Novozhilov, *Nestatsionarnoe Gorenje Tverdykh Raketnykh Topliv*, Nauka, Moscow, (1973). (Nonsteady Burning of Solid Rocket Propellants, Translation AFSC FTD-MT-24-317-74).
12. E.I.Maksimov and K.G.Shkadinsky, *Fiz. Goren. Vzryva.*, **7**, 454 - 458 (1971).
13. K.G.Shkadinsky and B.I.Khaikin, *Gorenje i Vzryv*, Nauka, Moscow, 104 - 109 (1972).
14. G.I.Barenblatt, Ya.B.Zeldovich and A.G.Istratov, *Zhurn. Prikl. Mekhaniki i Tekhn. Fiziki*, N° 4, 21 - 26 (1962).
15. G.M.Mackviladze and B.V.Novozhilov, *In: Teplo - Massoperenos*, (Proceedings of the IV All-Union Workshop on Heat- and Mass Transfer, Minsk), **2**, 454 (1972).
16. O.A.Borisova, V.B.Lidskii, M.N.Neigauz and B.V.Novozhilov, *Khim. Fizika*, **5**, 822 - 830 (1986).
17. O.A.Borisova, V.B.Lidskii, M.N.Neigauz and B.V.Novozhilov, *Fiz. Goren. Vzryva.*, N° 1, 81 - 84 (1990).

18. G.I.Sivashinsky, SIAM J. Appl. Math., **40**, 432 - 438 (1981).
19. V.A.Volpert, A.I.Volpert and A.G.Merzhanov, Dokl. Akad. Nauk SSSR, **262**, 642 - 645 (1982).
20. V.A.Volpert, A.I.Volpert and A.G.Merzhanov, Dokl. Akad. Nauk SSSR, **263**, 918 - 921 (1982).
21. V.A.Volpert, A.I.Volpert and A.G.Merzhanov, Fiz. Goren. Vzryva, **N° 4**, 69 - 72 (1983).
22. S.P.Radev and S.B.Shcherbak, Dokl. Akad. Nauk Bulgaria, **35**, 475 - 479 (1982).
23. S.B.Shcherbak, Fiz. Goren. Vzryva, **N° 5**, 9 - 12 (1983).
24. S.B.Shcherbak, Fiz. Goren. Vzryva, **N° 2**, 23 - 27 (1984).
25. S.B.Margolis, Prog. Energy and Combust.Sci., **17**, 135 - 162 (1991).
26. A.K.Filonenko, In Collected Edition: Combustion processes in chemical technology and metallurgy (edited by A.G.Merzhanov). Chernogolovka, 258 - 273 (1975).
27. A.K.Filonenko and V.I.Vershinnikov, Fiz. Goren. Vzryva, 353 - 362 (1975).
28. A.K.Filonenko, Archiwum termodynamiki i spalania, **6**, 23 - 35 (1975).
29. A.K.Filonenko, Fiz. Goren. Vzryva, **N° 6**, 41 - 45 (1991).
30. T.P.Ivleva, A.G.Merzhanov and K.G.Shkadinsky, Dokl. Akad. Nauk SSSR, **239**, 1086 - 1088 (1978).
31. T.P.Ivleva, A.G.Merzhanov and K.G.Shkadinsky, Fiz. Goren. Vzryva, **N° 2**, 3 - 10 (1980).
32. B.V.Novozhilov, Dokl. Russ.Akad. Nauk (in press, 1992)
33. Ya.B.Zeldovich, Zhurn. Ekspcr. i Teoret. Fiziki, **N° 11**, 159 - 168 (1941).
34. A.A.Zenin, A.G.Merzhanov and G.A.Nersisyan, Fiz. Goren. Vzryva, **N° 3**, 156 - 159 (1979).
35. A.A.Zenin, Pure and Appl.Chem., **62**, 889 - 897 (1990).
36. E.A.Butakova and A.G.Strunina, Fiz. Goren. Vzryva, **N° 1**, 71 - 73 (1985).